A SURVEY OF MONITORING AND ASSAY SYSTEMS FOR RELEASE OF METALS FROM RADIATION CONTROLLED AREAS AT LANL

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ABSTRACT

At Los Alamos National Laboratory (LANL), a recent effort in waste minimization has focused on scrap metal from radiological controlled areas (RCAs). In particular, scrap metal from RCAs needs to be dispositioned in a reasonable and cost effective manner. Recycling of DOE scrap metals from RCAs is currently under a self-imposed moratorium. Since recycling is not available and reuse is difficult, often metal waste from RCAs, which could otherwise be recycled, is disposed of as low-level waste. Estimates at LANL put the cost of low-level waste disposal at $550 to $4000 per cubic meter, depending on the type of waste and the disposal site. If the waste is mixed, the cost for treatment and disposal can be as high as $50,000 per cubic meter. Disposal of scrap metal as low-level waste uses up valuable space in the low-level waste disposal areas and requires transportation to the disposal site under Department of Transportation (DOT) regulations for low-level waste. In contrast, disposal as non-radioactive waste costs as little as $2 per cubic meter. While recycling is unavailable, disposing of the metal at an industrial waste site could be the best solution for this waste stream. A Green Is Clean (GIC) type verification program needs to be in place to provide the greatest assurance that the waste does not contain DOE added radioactivity. This paper is a review of available and emerging radiation monitoring and assay systems that could be used for scrap metal as part of the LANL GIC program.

BACKGROUND

A variety of radiation monitoring and assay systems have been developed across the DOE complex and for industrial purposes, both in the U.S. and abroad. Systems specific to LANL needs must address the challenge of monitoring for low levels of alpha emitting uranium and transuranic radionuclides. This is difficult and such measurement systems must be used as part of the larger institutional Green is Clean (GIC) program, including adequate acceptable knowledge, to do the best possible job of assuring that materials released from radiation areas are free of added radioactivity. Most existing systems focus either on lower density waste, contaminants with higher energy and intensity gamma rays, waste containing gamma emitting point sources, and waste with uniformly distributed radiation.

Setting up a system for GIC scrap metal verification at LANL will require that the following questions be answered

- What are the existing assay and monitoring systems for GIC applications?

- Can existing systems be adapted for this application?
- What are potential candidate technologies for further development?
- What recommendation can be made for a suitable system?

Each of the existing systems described is well known to the authors and has successfully addressed the specific issue of monitoring materials for radioactive contamination. In particular, the detection limits, administrative requirements, and mechanical aspects of this problem have been addressed for these systems. The prospective candidates for further development are in various stages of development, and a determination of suitability for further study has been made (using the CD-MOM as the baseline technology) and is presented in that section of the paper.

LANL has a number of cleanup operations in process and has significant amounts of metal scrap and waste in storage from previous cleanup operations. In addition to D&D-type cleanup operations, ongoing operations utilizing radioactive materials, such as the Plutonium Facility and the Neutron Science Center, produce metal scrap and waste as part of normal operations and upgrades to the facilities.

Metal scrap and waste from RCAs is currently being disposed of as low-level radioactive waste or stored for future reuse/recycling. Much of this waste could be determined to be free of radioactive contamination by a judicious use of process knowledge and a surveying and monitoring program combined with any required decontamination.

THE "GREEN IS CLEAN" & DECONTAMINATION PROGRAMS AT LANL

The Solid Waste Operations (SWO) group at LANL has a well-established GIC program (1) to disposition non-radioactive waste or recyclable materials from RCAs that is declared clean by generator acceptable knowledge (AK). LANL also has a Decontamination (Decon) Facility to decontaminate materials from RCAs. These programs are coordinated and supported by personnel from LANL's Environmental Stewardship Office (ESO), who perform a site-specific evaluation of applicable waste streams prior to acceptance for the GIC program.

The GIC non-destructive assay systems currently being used at SWO for verification that waste items are free of radioactive contamination include the Long Range Alpha Detection (LRAD), the Waste Assay for Nonradioactive Disposal (WAND), and the High Efficiency Radiation Counters for Ultimate Low Emission Sensitivity (HERCULES) systems. All three of the systems were constructed primarily for use with GIC waste from radiation areas with uranium and plutonium operations. LANL is also employing the Segmented Gate System (SGS) to segregate soil in D&D operations and a similar, smaller system to segregate clean and contaminated metal chips in the metal shops area. These systems are described in more detail below.

EXISTING TECHNOLOGIES

LRAD

Monitoring systems based on the LRAD principle detect the ions produced by the interaction of alpha particles with ambient air rather that the alpha particles themselves.(2) For greatest sensitivity filtered air is used to reduce radon in the assay chamber. The LRAD systems will not detect radiation on inaccessible (closed) surfaces, but they do detect radiation on surfaces available to the airflow, such as the inside of open pipes.

By definition, all ionizing radiation produces ions in air. The alpha particle deposits its energy within a few centimeters so LRAD-based detectors of reasonable size are primarily sensitive to alpha radiation.**(3)** The LRAD system sweeps the ions created in air by alpha contamination through ionization detectors to detect evidence of alpha contamination.

The system used at Los Alamos has an enclosure that is about the size and appearance of a large microwave oven. The inner dimensions of the sample chamber are 13 inches (33 cm) high by 22 inches (56 cm) wide by 16 inches (41 cm) deep.

The LRAD technology is the basis for several commercially available instruments from British Nuclear Fuels Instruments (BIL) and Thermo Eberline.(4,5) These instruments can be used to monitor irregular objects for alpha contamination - the differences are more in the size of acceptable objects than in the detection technology. LRAD instruments were designed specifically to facilitate the free release of metallic objects, but can also be used on non-metallic items.(6)

Other detectors with a similar appearance have been developed which utilize different detector technologies. Examples include large area gas proportional detectors with thin windows for alpha and beta contamination monitoring and plastic scintillation detectors for gamma contamination monitoring. Thermo Eberline produces several models of Automated Contamination Monitors (ATMs).

WAND and HERCULES

Both the WAND, a conveyor-based system, and HERCULES, a box counting system,**(7)** are highly sensitive gamma measurement systems optimized to detect very small quantities of common LANL radionuclides (e.g., Pu-239, Am-241, U-235, and U-238) in low-density waste. Both of the systems are prototypes developed at LANL that use a set of phoswich scintillation detectors in close proximity to the waste. These detectors have the capability of detecting activities below 3 pCi (0.1 Bq) per gram. These two systems will also detect high-energy betas and the entire spectrum of x-ray and gamma ray energies in low-density waste. In addition to detecting radiation, these systems are capable of identifying and quantifying a contaminant isotope.

The shredded low-density waste in the WAND system moves on a conveyor belt 2 inches below a bank of six detectors at a low rate of speed in an approximately 1 inch high by 12

inch wide stream. The HERCULES system consists of a bank of three detectors that screen low-density waste in 2 ft^3 cardboard boxes or in bags placed in a plastic 30-gallon drum on a turntable. The HERCULES can also be used to screen somewhat higher density waste that is homogeneous, such as metal chips.

The Segmented Gate System

The Segmented Gate System (SGS),(8) which is in use on soils and debris at LANL D&D sites, was developed by Thermo Nuclean. For proper operation, the soil and debris must be nominally less than 1.5 inches in diameter. This may require pre-sorting of the material prior to surveying. The depth of the soil or debris is adjusted based on the radionuclides of interest and the material characteristics. The primary radioactive contaminants of interest for the LANL projects are natural and depleted uranium, with a detection limit in the range of 50 pCi/g.

The SGS is a transportable gamma radiation detector system with a conveyor belt that operates at a constant speed and receives its feed from a hopper. It is designed to physically sort soil or debris with radioactive particles from clean soil or debris in the waste stream. There are pneumatic diversion gates at the end of the conveyor that divert contaminated material to a second conveyor and away from the non-contaminated material. This system utilizes two arrays of sodium iodide and beta detectors. When used for detecting gamma rays, the first set of detectors are thin (0.16-inch thick) sodium iodide detectors which are sensitive to gamma radiation from 15 keV to 200 keV. The second, thick array, consists of 2-inch thick sodium iodide detectors sensitive to gamma radiation from 150 keV to 1 MeV. One set of detectors may be replaced by a beta detector system that uses 100 cm^2 gas proportional counters. This system is well-suited for uranium, plutonium, americium, thorium, cesium or strontium contaminated soil and debris.

Chip Isolation System

Machining operations at LANL involving various metals including uranium create a mixture of radioactive and non-radioactive metal chips. Since the majority of the metal chips are non-radioactive, it is desirable to segregate the radioactive chips from the non-radioactive. The survey technique used in the past consisted of collecting the chips, spreading them onto a flat surface, and scanning the chips with a hand-held alpha/beta/gamma detector. The segregated non-radioactive metal chips were then sent through the LANL GIC assay equipment, where they are screened using the HERCULES system. HERCULES has a much lower limit of detection than the manual survey technique.

A new, more sensitive system for surveying the chips at the point of origin has been developed and tested.(9) This automated system prototype is called the Chip Isolation System, and consists of a chipper-shredder to reduce all of the metal chips to the same consistency with no chips being larger than 3 millimeters (one-eighth inch). The chipper-shredder dumps the chips onto a conveyor belt where they are spread evenly over the

surface of the belt. High-efficiency-plastic radiation detectors are positioned above the conveyor belt. The detectors are two large area plastic scintillators coupled to individual ratemeter electron monitors. The sensitivity of the system is 300 pCi of U-238. The detectors cover the width of the conveyor belt (18 inches) and they are 4.5 inches front to back. The conveyor belt speed is adjusted to a balance point between throughput and radiation detection level. The detector to source distance is approximately 1.5 inches.

Conveyor-Driven Medium Objects Monitor

Overhoff Technology Corporation has developed a line of products called U-ScanIt® radiation monitors. These monitors are designed to screen clean and low-level power plant waste for alpha, beta, and gamma radiation. Overhoff has recently deployed their Conveyor-Driven Medium Objects Monitor (CDMOM)(10) to Ontario Hydro to screen high-density waste leaving the nuclear power plant. The CDMOM detects contamination levels as low as 2 nCi. The detectors are arranged in an array around the conveyor belt. There are 12 gamma detectors and 16 alpha-beta detectors in the system. The gamma detectors are large-area plastic scintillators and the alpha-beta detectors are gas proportional counters. The detectors move automatically so they can be raised or lowered depending on the size of the items being monitored. This is accomplished using multiple low power laser beams to scan the incoming material. The system also automatically weighs the incoming material. This information is fed to the computer program and used to determine the level of contamination (if any). Under appropriate conditions, the minimum detectable activity for gamma is 5-10 nCi of cesium-137; for beta it is 5-10 nCi of technicium-99.

Scrap Metal Recycling and Portal Monitors

Vehicle portal monitors designed specifically to detect special nuclear material (SNM) such as plutonium and uranium are in operation at LANL at the solid waste Material Recycling Facility (MRF) and at entrances to SNM processing areas.(11) The Los Alamos County Landfill also employs such a portal monitor. These monitors utilize plastic scintillators as the means of detecting radioactive material. Other portal monitors have been designed that use neutron detection based ^{3}He detector systems. Several basic designs are discussed in reference #11.

In the MRF facility, personnel inspect, sort, and segregate solid waste from (non-radioactive) LANL operations into recyclable materials, prohibited materials, and sanitary landfill waste. The sorted wastes are compacted and bailed. As trucks enter the facility, they pass through a portal monitor manufactured by TSA Systems, Ltd.(12) as a check that the facility is not receiving any waste contaminated with radioactive material. The portal monitor consists of two self-contained pillars placed on either side of the vehicle path. Each pillar contains two plastic scintillator detectors, an occupancy detector, and associated electronics. In other systems, the detectors completely surround the vehicle path. This TSA system was specifically built to detect U-235 and Pu-239. The sensitivity for this system is specified in terms of cesium as 7 μCi of cesium-137 at a

vehicle speed of 5 miles per hour at 20 foot pillar spacing in a 20 mR/hour background. Other systems minimum detectable activity is specified as low as 0.3 μCi of cesium-137.

Direct and Indirect Survey Techniques

Traditionally, both direct and indirect health physics surveys have been used to determine whether an item leaving an RCA can be considered free of radioactivity. These techniques are used at the LANL Decon facility to determine whether metal and other materials from RCAs are uncontaminated. Typically, indirect smears (for removable contamination) as well as direct monitoring instruments are used for this purpose. The Decon facility utilizes both large area and 100 cm^2 area smears; these are measured with a Ludlum ESP sample counter. The sample counters can detect contamination on the smears of less than 10 disintegrations per minute (dpm) alpha. Direct surveys are accomplished using hand held alpha/beta/gamma contamination detectors manufactured by Berthold.(13) The Decon facility has been used to clear recycle metal scrap in the past (prior to the moratorium). It is a labor-intensive, and therefore, expensive activity and throughput is limited.

Effective Automatic Monitoring for Transuranics

Ideally, a sensitive, automatic system could be built specifically to verify that metal scrap from RCAs that handle or store alpha-emitting uranium and transuranic radionuclides is free of DOE-added radioactivity. This could include scrap metal that has interior surfaces or thick walls.

Plutonium-239, the primary transuranic radionuclide of interest, is an alpha emitter that also produces a large number of x-rays and gamma rays. The alpha particles will not penetrate metal and have a very short range in air. The x-rays are low-energy and will not penetrate very far through metal. The gamma rays have a higher energy than the x-rays, but extremely low yield (approximately 10^{-5}).

Plutonium-239 is usually found in conjunction with other plutonium isotopes (238, 240, 241, and 242) in smaller quantities. All of these plutonium isotopes emit alpha particles (although the plutonium 241 alpha has a very low yield of about 10^{-5} ;it is not practical to use it for detection). The most common grade of plutonium contains approximately 94% plutonium-239,6% plutonium-240and very small amounts of the other plutonium isotopes. Plutonium-240 has a few low-energy gamma rays with yields in the 10^{-5} – 10^{-7} region; however, it is a strong spontaneous neutron emitter, which makes it a good candidate for neutron detection. Plutonium-241 emits low energy beta radiation and also generates low energy and low yield gamma rays. Plutonium-242 emits a few gamma rays, but has a very low abundance in the typical plutonium isotopic mixture. The plutonium isotopic mixture also produces decay products such as –americium-241, uranium-237 and neptunium-237. The americium-241 has a very strong low energy gamma signal and several high- energy gammas with low yield (in the 10^{-6} region). The uranium-237 and neptunium-237 are present in relatively small quantities.

Uranium-235, the primary uranium isotope of interest, is also an alpha emitter. It is found in varying proportions with uranium-238 and (usually) minor amounts of other uranium isotopes. Uranium suffers from many of the same detection problems that plutonium does, however, the intensity of radiation from U-235 is even less than Pu-239, and U-238 has an even lower intensity than U-235. Table I compares the number of gamma rays from plutonium and uranium.(14) The spontaneous fissioning of uranium is rare compared to the rate in Pu-240, but U-235 can be excited and assayed using active neutron techniques. U-235 can be detected using gamma detection technology if a sufficient quantity is present, but like plutonium, U-235 emits low energy, low intensity gamma rays. U-238 is normally detected by gamma rays from its equilibrium daughter product protactinium-234m, which are high energy, but again, low intensity, gamma rays.

Table I. Primary gamma rays for major radionuclides of interest

Isotope	Energy (keV)	Activity (γ/g-s)	Mean Free Path (mm), High-Z (ρ=10 g/cm^3)	Mean Free Path (mm), Low-Z (ρ=1 g/cm^3)
U235	185.7	4.32×10^4	0.69	80
U238[a]	1001.0	7.34×10^1	13.30	159
Pu239	129.3	1.44×10^5	0.27	71
Am241	59.5	4.54×10^{10}	0.14	38

[a]From U238 daughter Pa234m. Equilibrium assumed.

POTENTIAL CANDIDATES FOR FUTURE RESEARCH AND DEVELOPMENT

The limitations of the radioactive decay outlined above combined with the matrix properties of scrap metal from D&D and ongoing operations in RCAs present a unique problem. In an attempt to design an effective monitoring system that would go beyond the existing radiation assay and monitoring systems described above, a literature search was undertaken with several objectives:

- looking beyond conventional thinking about instrumentation for this purpose,
- conceiving of a system with an innovative design that is sensitive to a greater proportion of the matrix material surfaces, and
- combining several different approaches into one system.

Many of the standard instruments that may be applicable, in some modified or complementary form, to this problem have already been discussed section above. If the measurement system will only be required to detect that radioactive contamination is present; not identify or quantify it, potential candidates for application to this monitoring issue include:

- Pulsed Gamma Neutron Activation Analysis(15) (PGNAA) technique that is used by Westinghouse to detect heavy metals (including uranium),
- imaging devices, such as RADCAM(16)
- a device adapted from liquid flow monitoring, or gaseous flow monitoring, such as a high pressure scintillation detector,(17)
- ambient air ionization monitors other than the LRAD,
- membranes to transport small detectors,
- phosphor technology,(18) and
- modified dosimetry technology.

The PGNAA and similar techniques such as the Pulsed Fast/Thermal Neutron Analysis(19) (PFTNA) or Prompt Gamma Activation Analysis(20) (PGAA), have been used primarily to detect contaminants (e.g., RCRA metals) and, more recently, transport of contraband materials. These systems utilize a source of neutrons that excite a target nucleus. The excited nucleus then radioactively decays by the emission of prompt and delayed gamma rays and other radiation such as beta particles. The emitted radiation is characteristic for the particular target nucleus and can be used to identify it. This technology has been used successfully in both laboratory and field applications, but has not been applied to the detection of small amounts of plutonium in a field operation.

Imaging devices have been developed for low intensity gamma-ray sources. For example, the RADCAM, a product of Radiation Monitoring Devices, Inc., uses a position sensitive photomultiplier tube coupled to a high light output CsI(Na) scintillation crystal. After processing, the radiation image is overlaid on a video picture captured by a high-resolution charge coupled device. While radiation imaging may not be practical or desirable for this application, the methods of reducing background radiation and noise employed in the technology may be very useful. Concepts used in the Poissonian-Type Radiation Imager(21) technology being developed for high-energy physics experiments at CERN, using fibers to create a larger scintillation detector, may provide a route to developing larger and more sensitive scintillation detectors that could be applied to this problem.

MELTILEX™ is a meltable scintillator that has been used as a thin coating layer on the inner surface of a bundle of thermoplastic, transparent tubes.(22) This bundle has been used to detect alpha particles in solutions. The detection limits are as low as 0.15 nCi/ml in a liquid flow. A high-pressure scintillation detector has been used to detect low levels of radioactive gases released into the atmosphere from nuclear power plants, fuel reprocessing plants, and nuclear weapons tests. This detector measures beta and internal conversion electron decays of fission-produced radionuclides. The high-pressure feature of this detector allows for the concentration of electrons to be detected and results in a higher efficiency detector. Gas flow rates of 5-15 liters per minute have been achieved.

Ambient air ionization monitors other than the LRAD have been developed for application to monitoring for alpha contamination. The E-PERM® Alpha Surface Monitor,(23) for example, uses an electret ion chamber (EIC) to measure alpha radiation on surfaces of materials. The electret is a positively charged piece of Teflon® that

attracts electrons from air ionized by alpha particles. The electret is removed from the chamber and read on an external voltage reader. A change in the electret voltage indicates the presence of alpha contamination and the voltage drop and exposure time are converted to contamination levels.

Thin membranes propagated through pipe runs using air pressure have been used as sensor deployment devices. The Pipe Explorer® system(24) built by Science and Engineering Associates, Inc., utilizes this technology by remotely emplacing scintillators inside complex pipe runs as small as two inches in diameter and as long as 250 feet. The scintillators are directly in contact with the inner pipe walls. A photodetector is towed inside, producing a log of the alpha contamination in the pipe. A system such as this might be adapted in a practical manner for the current problem.

Storage photostimulable phosphor (SPP) technology has been developed for radiation detection in environmental samples. This technology exhibits high sensitivity for all types of radiation and the response is linear over a wide dynamic range. These sensors can be used as large area detectors, with an active area up to 35x43 cm (14x17 inches). The sensors are exposed to the area to be surveyed; energy is trapped in the sensor and later released when the sensor is scanned by a laser beam in a reader unit. A digital image of the radiation field on the sensor is produced, producing a "map" of the radiation detected. The sensors are reusable and can be calibrated to calculate total activity.(25)

Finally, technology traditionally thought of as dosimeter technology might be adapted for use in this arena. Dosimetry technology has been applied to various commercial D&D projects and is a long-term initiative. Along this same vein, superheated drop detectors (SDD) have been under development for several years. Sensitive neutron dosimeters have been fabricated using this technology.(26) The response of the SDD is proportional to the neutron dose.

TECHNOLOGY COMPARISON

A qualitative comparison of the technologies described in this paper is presented in Table II below. The Conveyor-Driven Medium Objects Monitor was chosen as the baseline technology to evaluate the other technologies. It is already in operation in a manner that duplicates the physical requirements of the current problem (though not the radiological requirements). The "Level of Development" column in this table only refers to instrument development and not to engineering system development for system automation, e.g., the LRAD may require a greater degree development for the automation function than many of the other technologies, but the instrument itself is well-developed.

Table II. A qualitative comparison of technologies

Technology Type	Suitable for Metals?	Suitable for Auto-mation?	Sensitivity Compared to CDMOM	Throughput Compared to CDMOM	Level of Develop-ment Required	Further Study Warranted?
LRAD	Yes	Yes	High	Low	Low	Yes
WAND	No	Yes	High	Low	N/A	No
HERCULES	Some	Yes	High	Low	N/A	No
Segmented Gate System	Some	Yes	High	Similar	Low	Yes
Chip Isolation System	Some	Yes	High	Similar	Low	Yes
CDMOM	Yes	Yes	CDMOM	CDMOM	Low	Yes
Portal Monitors	Yes	Yes	Low	Similar	High	No
Survey Techniques	Yes	No	High	Low	Low	No
Activation Analysis	Yes	Yes	Low	Low	Low	No
Imaging Devices	Yes	Yes	Lower	Similar	Low	No
High Pressure Scintillation	Maybe	No	High	Low	High	Maybe
Air Ionization Monitors (not LRAD)	Yes	Maybe	High	Low	Low	Yes
Thin Membranes	Yes	Maybe	High	Low	Medium	Yes
Phosphor Technology	Yes	Maybe	High	Low	Medium	Yes
Modified Dosimetry Technology	Yes	Maybe	High	Low	Medium	Yes
Neutron Technology	Yes	Yes	Low	Low	Low	No

CONCLUSION

Several of the technologies described in this paper are candidates for further study in recommending a verification system for uncontaminated metals from RCAs. In particular, those that are already highly developed, are suitable for metals, and are suitable for automation, will receive further attention. Technologies listed in the table above as having sensitivities lower than the CDMOM were automatically eliminated from further consideration. Similarly, systems that were obviously not suited for metals

were eliminated, and the HERCULES was eliminated because it is only suitable to very low density metals.

As evidenced by the discussion of existing systems above, the mechanics of a conveyor type system for verifying the absence of uranium and transuranic radionuclide contamination on metal scrap is a solvable engineering challenge. The greater challenge comes in fielding a system that will adequately perform the verification task in a timely and cost effective manner and that will also satisfy LANL and DOE management and outside regulators. Continued efforts are being undertaken to address this problem based on the technology review outlined in this paper.

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